11

Vegetation and Foodstuff Monitoring

S. Ring Peterson

Introduction

Lawrence Livermore National Laboratory has a vegetation and foodstuff monitoring program to comply with U.S. Department of Energy (DOE) guidance. This guidance (U.S. DOE 1991) states that periodic sampling and analysis of vegetation should be performed to determine if there is measurable, long-term buildup of radionuclides in the terrestrial environment.

Tritium is the nuclide of major interest in the LLNL vegetation and foodstuff monitoring program. LLNL has historically released tritium to the air accidentally and during routine operations, and tritium is the only radionuclide released from LLNL activities that occurs in detectable concentrations in vegetation and foodstuff. Tritium moves through the food chain as tritiated water and can be rapidly assimilated into plant water and then incorporated into the organic matter of plants through photosynthesis. It can contribute to human radiation dose if it is inhaled, absorbed through the skin, or ingested via vegetables or via milk and meat from animals that have eaten tritiated vegetation.

LLNL has been monitoring tritium in vegetation to some extent since 1966 and has performed vegetation sampling in the vicinity of the Livermore site and Site 300 as part of a continuing monitoring program since 1971. The monitoring program is designed to measure changes in the environmental levels of radioactivity, to evaluate the environmental effect of LLNL operations, and to calculate potential human doses from radionuclides in the food chain.

In 1977, LLNL added wine to the LLNL monitoring program. Wine is now the most important agricultural product in the Livermore Valley, representing an approximately $80-million annual industry, based on sales. Although the tritium concentrations in all wines are low, the sampling data indicate that Livermore Valley wines contain statistically more tritium than do wines from other California wine-producing regions.

In the past, other foodstuffs (cow milk, goat milk, and honey) leading to potential dose were also monitored for tritium. At present, however, only tritium concentrations in vegetation and wine are used to assess potential ingestion dose from tritium emitted during LLNL operations. During 2000, LLNL collected and analyzed samples of herbaceous vegetation and wine. Potential human doses from these foodstuffs were calculated using the monitoring data and the dose models presented in Appendix A. In addition, as part of a continuing study, LLNL determined the potential tritium dose to the maximally exposed individual from a pine tree at the Livermore site. This tree serves as a diffuse source of tritium because it loses tritium to the air through evapotranspiration of tritium-contaminated water in the root zone. The dose was calculated using the U.S. Environmental Protection Agency (EPA) model CAP88-PC.

Methods

The methods used for monitoring vegetation and wine are presented briefly in the following sections and in more detail in the Data Supplement. All vegetation and wine sampling was conducted according to written and approved standardized procedures (Tate et al. 1999).

Vegetation

LLNL staff collected vegetation samples, usually annual grasses, quarterly from 22 fixed locations in the Livermore Valley, San Joaquin County, and Site 300. The samples were then analyzed for tritium.

Location maps are provided in **Figures 11-1** and **11-2**. Sample locations were selected to represent vegetation from locations near LLNL that can be affected by LLNL operations, background locations where vegetation is unlikely to be affected by LLNL operations, and areas of known or suspected LLNL-induced contamination. In 2000, the location COHO, at Site 300, replaced PRIM after the first quarter; all other sampling locations were the same as those in 1999.

Wine

Three types of wine samples were collected and analyzed for tritium concentrations: wine produced from grapes grown in the Livermore Valley, wine produced from grapes grown in California outside the Livermore Valley, and wine produced from grapes grown in Europe (France, Germany, and Italy). The wines were purchased from local retailers to represent what the general public could buy and drink during 2000.

Data from the analysis of tritium in wine can be used to estimate the potential tritium dose received by consumers during the year of purchase. However, because wines purchased in 2000 are from grapes that were harvested in 1997, 1998, and 1999, the 2000 sampling data cannot be used to indicate how LLNL's operations affected concentrations of tritium in wines produced from grapes grown in 2000. To analyze trends and help determine the impact of LLNL operations on tritium in wine for the year of harvest, LLNL corrects the wine concentrations for radiological decay that has occurred between the approximate date of the grape harvest and the date when the wine was analyzed in the laboratory. Comparisons can then be made of wine concentrations that represent the year when the grapes were exposed to the tritium.

Results

The results of vegetation monitoring for the Livermore site and Site 300 and the results of wine monitoring are presented in the following sections.

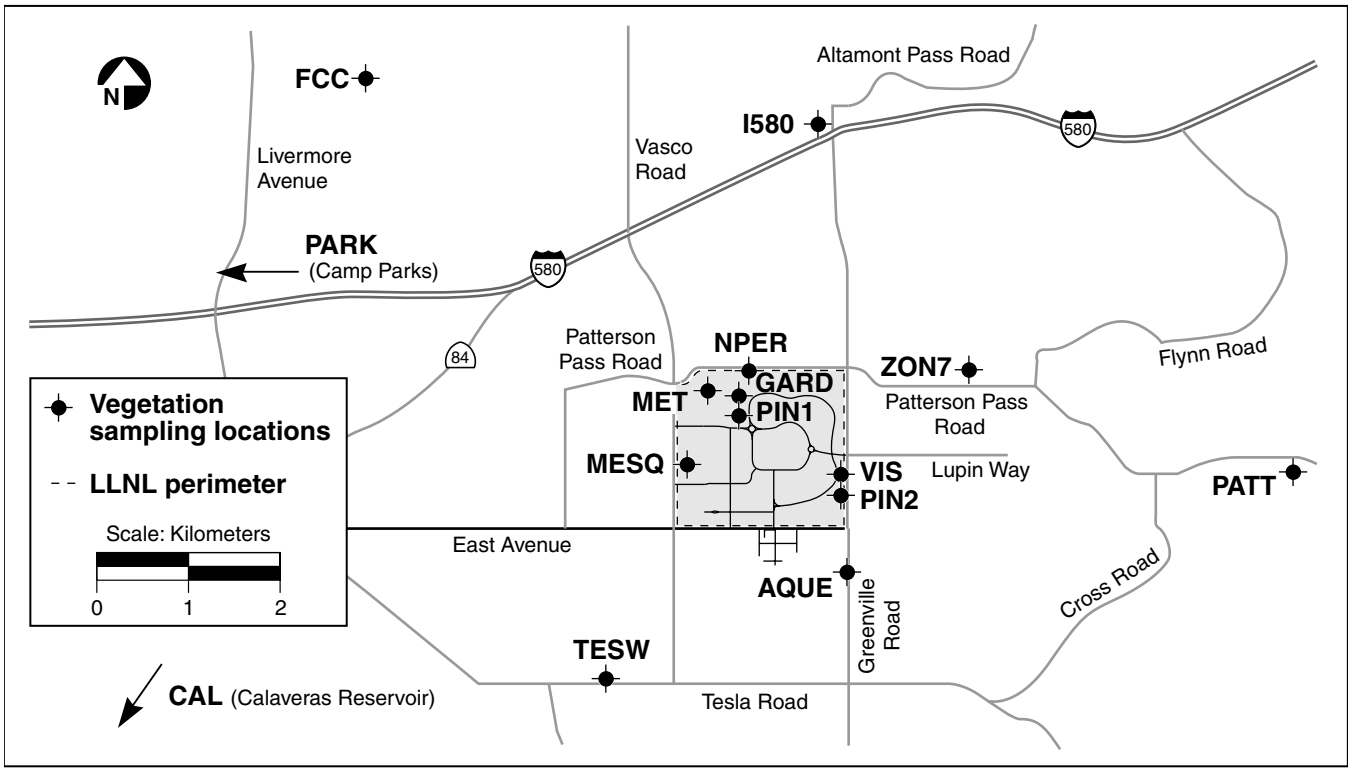

Figure 11-1. Livermore site and Livermore Valley vegetation sampling locations, 2000

Livermore Site

Vegetation

The Livermore site and Livermore Valley vegetation locations are put into four groups for statistical evaluation:

- **Near:** locations within 1 km of the Livermore site perimeter. Near locations include AQUE, GARD, MESQ, NPER, MET, PIN2, and VIS.

- **Intermediate:** locations in the Livermore Valley 1–5 km from the Livermore site perimeter that are often downwind and, thus, potentially under the influence of tritium releases at the site. The intermediate locations are I580, PATT, TESW, and ZON7.

- **Far:** locations unlikely to be affected by LLNL operations. One background location (CAL) is more than 25 km away. The other two (FCC and PARK), although in the Livermore Valley, are unlikely to be affected by LLNL operations because they are more than 5 km from the Livermore site and are generally upwind.

- **PIN1:** location of a pine tree rooted in an area of known tritium contamination on the Livermore site.

Table 11-1 shows summary tritium data for vegetation collected for the LLNL vegetation monitoring program in 2000 (individual sampling values are presented in the Data Supplement of this report). **Figure 11-3** shows the 2000 medians

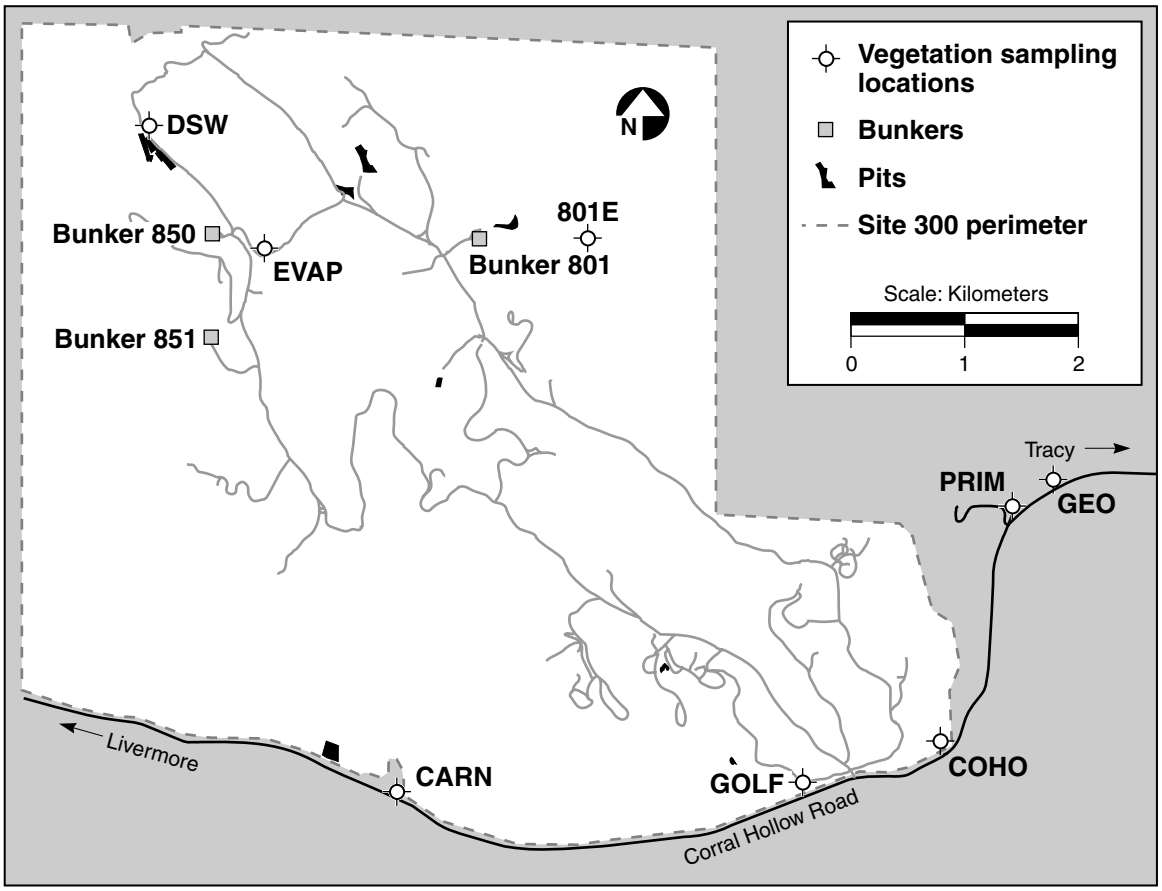

Figure 11-2. Site 300 vegetation sampling locations, 2000

of the tritium concentrations for PIN1, Near, Intermediate, and Far Livermore locations as a continuation of historic median concentrations from 1971 to 1999. For 1998 and 1999, the medians for the Far locations were negative; therefore, because negative numbers cannot be plotted on a logarithmic scale, the lowest positive measured concentrations were plotted instead. Although the concentration in Far vegetation appears to drop by about a factor of 10 in 1998 and to rise sharply in 2000, it is highly unlikely that these apparent changes represent concentrations different from recent preceding years. The apparent difference in the Intermediate and Far observations since 1998 is caused by a change in how the analytical laboratory reported concentrations less than the detection limit. All Far vegetation samples were below the detection limit of the analysis in 1999 and 2000.

For 2000, the data for tritium in vegetation were compared using Scheffé's *F* and Games/Howell multiple comparisons (Scheffé 1953; Games and Howell 1976). These tests are the most appropriate tests for these distributions of data. Once again, as in 1999, the Near group was not found to be significantly different from the Intermediate and Far groups. This is due to many low observed tritium concentrations in the Near samples. For example, the first quarter sample for VIS was below the detection limits for the first time since sampling began there in 1991 (see Table 11-1 in the Data

Table 11-1. Concentrations of tritium in plant water (Bq/L), 2000

Location [a]	Detection Frequency [b]	Median	Inter-quartile Range	Maximum	Dose (nSv/y)[c]	
					Median	Maximum
Livermore site near locations [d]	23/28	2.4	4.0	12	12	59
Livermore site PIN1 [e]	4/4	59	68	240	0.0045[f]	0.018[f]
Livermore site intermediate locations	6/16	1.2	2.1	8.3	5.9	41
Livermore site far locations	0/12	0.48	1.6	1.9	2.4	9.3
Location DSW at Site 300 [e]	3/4	5.8	320	1300	28	6400
Location EVAP at Site 300 [e]	3/4	160	170	290	780	1400
All other locations at Site 300	2/20	0.21	1.0	1.8	1.0	8.8

Note: Radioactivities are reported as the measured concentration and an uncertainty ($\pm 2\sigma$ counting error). If the concentration is less than or equal to the uncertainty the result is considered to be a nondetection. See Chapter 14.

a See **Figures 11-1** and **11-2** for sampling locations.

b Detection frequency means the fraction of samples taken having a measured value above the detection limit.

c Ingestion dose is calculated based on conservative assumptions that an adult's diet is exclusively vegetables with this tritium concentration and that meat and milk are derived from livestock fed on grasses with the same concentration of tritium. See Appendix A. Note that doses are reported in nSv/y (1×10^{-9} Sv/y) rather than the μSv/y (1×10^{-6} Sv/y) of earlier years.

d Includes PIN2; plant water concentrations are similar among plant types

e Sampling location in area of known contamination

f For this dose calculation PIN1 is treated as a diffuse source of tritium (since pine needles are not eaten by human beings). Dose calculated using CAP88-PC is to the maximally exposed individual (see Chapter 13).

Supplement). The highest median tritium results for individual vegetation sampling locations were found at the Near locations, NPER and VIS, both of which were lower than in 1999.

In 1997 PIN1, a pine tree growing in a known area of tritium contamination at the Livermore site, was monitored on a monthly basis to estimate emissions for compliance with National Emissions Standards for Hazardous Air Pollutants (NESHAPs). In 1998, the tree sampling was coordinated with the quarterly vegetation sampling. Since 1998, NESHAPs calculations to the maximally exposed individual (MEI) have been based on quarterly observations. To assess the contribution of soil water tritium to PIN1, LLNL also sampled a second tree (PIN2) which is not growing in tritium-contaminated soil. Concentrations of tritium in PIN2, like in all other vegetation sampled near the Livermore site, are from air and soil water in quasi-equilibrium with air. When samples from PIN1 were compared with samples from each Near location for 2000 using Scheffé's *F* procedure, concentrations of tritium in PIN1 were found to be significantly higher than concentrations at all other locations at the 5% significance level.

Wine

The results from the 2000 wine tritium analyses are shown in **Table 11-2**. Tritium concentrations are within the range of those reported in previous years and remain low in wines from all areas. The data for the 2000 sampling year were analyzed using Scheffé's *F* and Games/Howell multiple comparisons. The results of the comparisons are the same as in previous years. Both analyses show that the tritium concentrations of Livermore Valley wines are higher than those of the six California

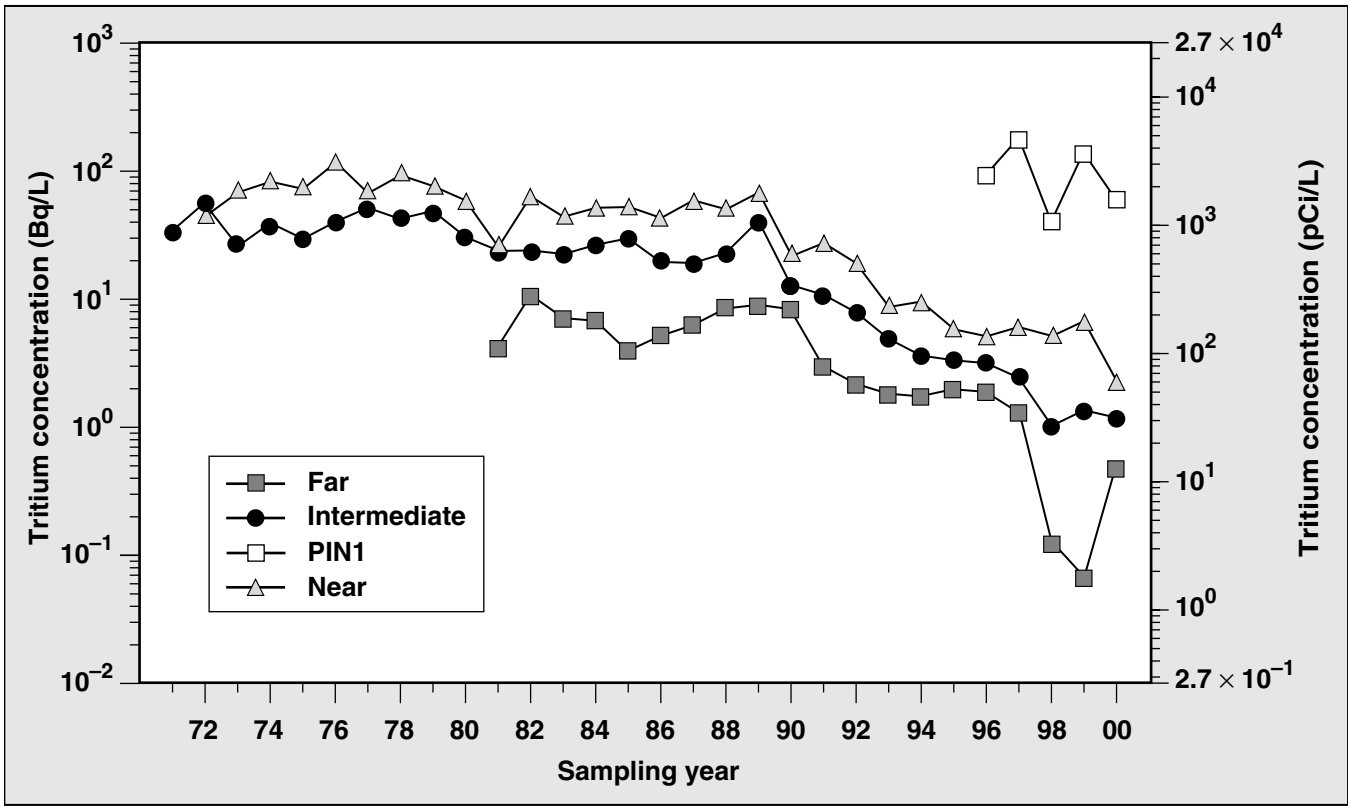

Figure 11-3. Median tritium concentrations in Livermore site and Livermore Valley plant water samples, 1971–2000 (For Far vegetation for 1998 and 1999, the values are the lowest positive.)

Table 11-2. Tritium in retail wine (Bq/L), 2000[a]

Region	Detection[b] frequency	Median	Interquartile range	Mean	Maximum	Dose[c] nSv/y
Livermore Valley	12/12	1.80	0.523	1.75	2.62	1.6
California	6/6	0.424	0.0340	0.449	0.584	0.40
Europe	4/4	1.15	0.615	1.18	1.61	1.1

Note: Radioactivities are reported as the measured concentration and an uncertainty ($\pm 2\sigma$ counting error). If the concentration is less than or equal to the uncertainty, the result is considered to be a nondetection. See Chapter 14.

a Wines from a variety of vintages were purchased and analyzed in 2000. The concentrations reported are those at the time the bottle was opened.

b Detection frequency means the fraction of samples taken having a measured value above the detection limit.

c This dose is calculated from assumption of drinking 52 L wine/year and using the mean concentration of sampled wines.

wines at the 5% significance level. The Scheffé's *F* test, which can be used when the number of samples is fewer than six, also demonstrated that the California wines sampled have significantly lower tritium concentrations than the European wines sampled.

There is remarkably little variability in Livermore Valley wines collected for 2000, although the vintage years represented are 1997, 1998, and 1999.

Concentrations of tritium in wine corrected to vintage year are plotted in **Figure 11-4**. The downward trend seen in wines from all locations between 1982 and 1999 exhibits approximately a five-year half-life. The radiological half-life of tritium is 12.32 years. An environmental half-life shorter than the radiological half-life would be expected for tritium in Livermore Valley wines, since concentrations in grapes are driven by emissions from LLNL, and emissions have dropped since 1988, with a significant reduction after 1992 (see Chapter 4). What is surprising is the observation that Californian and European wines exhibit a similar half-life. The locations where the grapes were grown and the corresponding driving concentrations of tritium (e.g., fallout for California, unknown for Europe) only seem to affect the magnitude of the tritium concentrations, not the rate of loss from the environment. This observed loss-rate of tritium from wines is most likely due to the transfer of tritium to the oceans, which are a sink for tritium, and is similar to the loss-rate of tritium from precipitation observed in similar years after the cessation of bomb tests (Östlund, 1973; Momoshima et al. 1991).

The 1996 Livermore Valley wines were significantly higher in tritium than those from both 1997 and 1998 using both Scheffé's *F* procedure and the Games/Howell test. Data from 1999 could not be included because the sample comprised only two

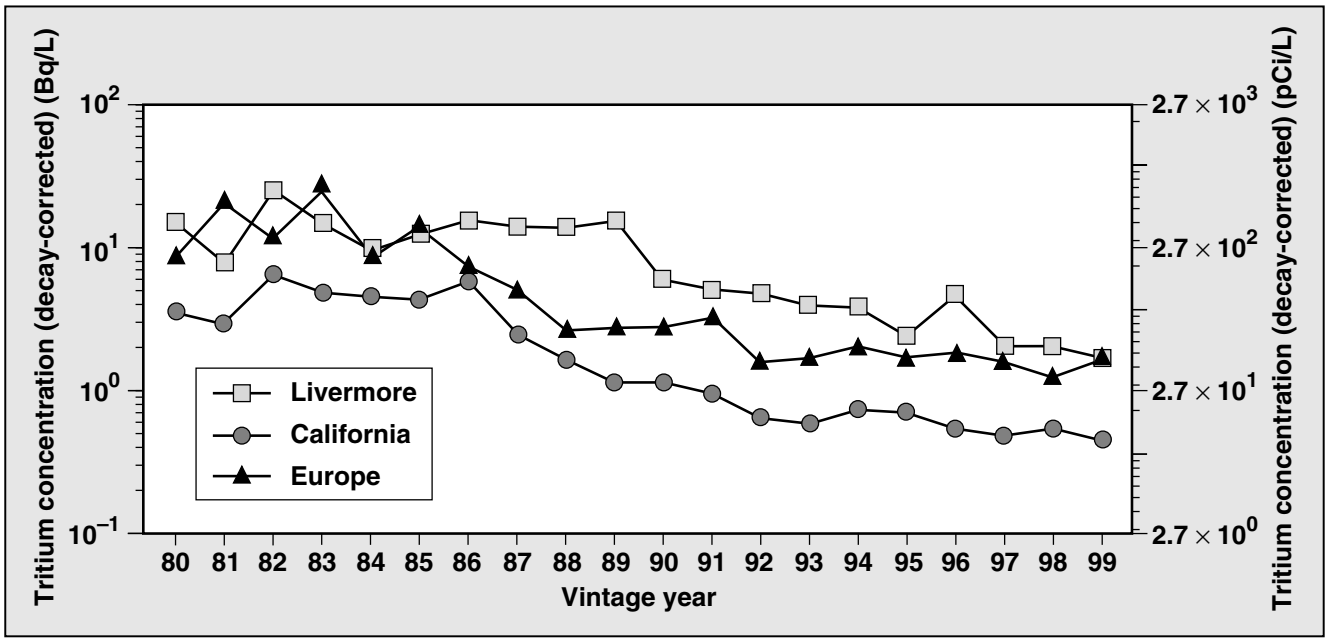

Figure 11-4. Mean tritium concentrations in retail wines decay-corrected from the sampling year to the vintage year

bottles. As mentioned, wines are sampled randomly. Quite by chance, the 1996 wines unequally represent vineyards close to LLNL and therefore exhibited higher values.

Site 300

Vegetation

There are seven monitoring locations for vegetation at Site 300. Of these, five (CARN, GOLF, GEO, 801E, and PRIM/COHO) detect changes in atmospheric tritium concentrations. Vegetation from locations DSW and EVAP grows in areas of known groundwater contamination. Plants can take up tritiated water from two sources: air moisture and soil moisture. When soil water available to the plant is contaminated with tritium and there is little tritium in the air moisture, the concentrations in the plant water will be somewhat lower than those of soil water, but will be much higher than concentrations in air moisture.

Table 11-1 shows summary tritium data for vegetation collected at Site 300 during 2000. Historic values for tritium at Site 300 sampling locations are shown in Figure 11-5. Of the seven sampling locations at Site 300, five yielded results in 2000 at or near the detection limits. Two locations, EVAP and DSW, yielded results above detection limits. EVAP's median tritium concentration is similar to that of 1999. DSW's median value is similar to recent years (excepting 1998). The extremely low concentrations from 1998 to the present for locations other than DSW and EVAP are caused by a change in how the analytical laboratory reported concentrations lower than detection limit and do not reflect a real difference in concentrations.

The highest tritium result (1300 Bq/L) for a single vegetation sample occurred at location DSW (see Table 11-1). This sampling location is adjacent to a landfill area that contains debris contaminated with tritium from past experiments. Tritium concentrations in vegetation are also above background levels at location EVAP, which is near a spring where groundwater flows near the surface and evaporates. Groundwater near EVAP is contaminated with tritium from Pit 3, Pit 5, and the firing table at Building 850. The DSW and EVAP locations are both within the East and West Firing Area (EFA/WFA) Study Area of the Comprehensive Environmental Response, Compensation and Liability Act (CERCLA) environmental restoration study areas (see Chapter 8, Groundwater Investigation and Remediation). Tritium is one of the contaminants of concern in this area. Relatively high concentrations of tritium in plants at DSW and EVAP are observed only sporadically when conditions result in contaminated soil water being translocated by vegetation. Evaluation of the 2000 data for Site 300 using Scheffé's *F* procedure yielded no significant differences among the various sampling locations, a result of the high variability of the data and the low number of data points.

Environmental Impact

In 2000, the environmental impacts of LLNL operations on vegetation and wine, presented below, were small.

Livermore Site

LLNL impacts on vegetation in the Livermore Valley remained minimal in 2000. The effective dose equivalents shown in Table 11-1 were derived using the dose conversion factors provided by DOE (U.S. DOE 1988) and the dose pathway model from U.S. Nuclear Regulatory Commission (NRC) Regulatory Guide 1.109 (U.S. NRC 1977). Appendix A provides a detailed discussion of dose calculation methods. The dose from ingested tritium is based on the conservative assumptions that an adult's diet (Table A-2, NRC maximum) consists exclusively of leafy vegetables

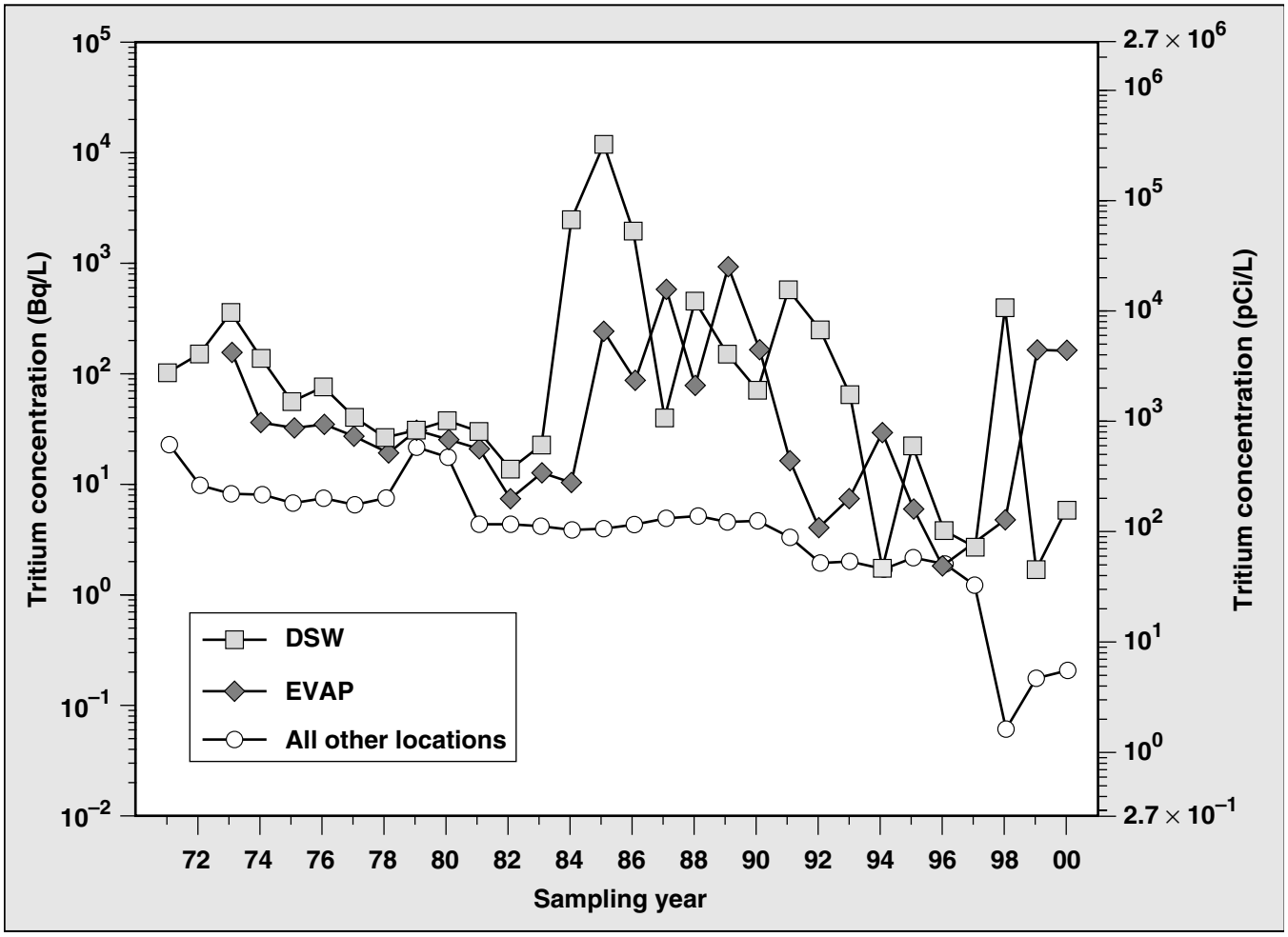

Figure 11-5. Median tritium concentrations in plant water at Site 300 sampling locations, 1971–2000. When the median is negative (e.g., all other locations for 1998 and 1999), the lowest positive concentration has been substituted.

with the measured tritium concentrations, as well as meat and milk from livestock fed on grasses with the same concentrations. In actuality, the vegetables consumed by an adult contain tritium at lower levels than those reported because most vegetables are imported from other areas. Similarly, tritium concentrations in food consumed by local livestock are at or below the concentrations in vegetation measured at the Intermediate and the Far locations. Nevertheless, based on these extremely conservative assumptions, the maximum potential dose from ingestion of affected vegetation for 2000 for the Livermore Valley is 58 nSv/y (0.058 µSv or (0.0058 mrem).

Doses are calculated based on measured tritium in plant water without considering the contribution of organically bound tritium (OBT). Dose conversion factors of 1.8×10^{-11} Sv/Bq for tritium in the plant or animal water (HTO) and 4.2×10^{-11} Sv/Bq for OBT have been established by the International Commission on Radiological Protection

(1996). These show the relative importance of ingested HTO and OBT to dose. When vegetables are ingested, the dose from the HTO contribution is greater than the dose from the OBT contribution because the fraction of the vegetable that is organic matter is quite small (10–25%). For example, about 10% of the ingestion dose from leafy vegetables (about 10% dry matter) is contributed by OBT. OBT becomes increasingly important to dose when the fraction of dry matter increases. Pork, for example, has a dry-matter content of about 30–50% (Ciba-Geigy Ltd. 1981), and the resulting ingestion dose from pork is about half from OBT and half from HTO. The OBT in grain, which is 88% dry matter, contributes nearly 90% of the dose from ingested grain. Given the different fractions of OBT in different foods, the importance of OBT to ingestion dose depends on what quantities of what kinds of foods are consumed. An extremely unlikely diet very high in OBT would, at most, give an OBT contribution to dose equal to that of HTO. Thus, conservatively, the maximum total tritium dose from ingestion of vegetables and foodstuffs from the Livermore Valley for 2000 could be 120 nSv (0.12 µSv or 0.012 mrem), which is more than a factor of eight below the estimate for 1999 and well below any level of concern.

The dose values for PIN1 shown in **Table 11-1** were calculated in a different manner from those for edible vegetation, because it is unreasonable to assume that any person is directly ingesting pine needles. The pine tree is treated as a diffuse source of tritium to the atmosphere via the contaminated transpirational stream. LLNL used an estimated tritium transpiration rate from the tree as input data to the EPA regulatory model CAP88-PC. LLNL modeled air dispersion of the transpired tritium and calculated a resulting dose from inhalation, skin absorption, and potential ingestion from air concentrations at the location of the maximally exposed individual. This total dose is based on the conservative assumptions that 100% of the individual's time is spent at this location and that his/her diet consists exclusively of vegetables with the measured tritium concentration and meat from livestock fed on grasses with the same concentration. The resulting maximum dose for PIN1 of 0.018 nSv/y (1.8×10^{-5} µSv or 1.8×10^{-6} mrem) is considerably lower than ingestion doses calculated directly from measured concentrations in vegetation because the tree is only an indirect source of air/vegetation contamination.

No health standards exist for radionuclides in wine. However, all the wine tritium levels were far below drinking water standards. In fact, even the highest detected Livermore Valley value (2.3 Bq/L or 62 pCi/L) represents only 0.3% of the California drinking water standard (740 Bq/L or 20,000 pCi/L). Doses from wine consumption can be calculated according to methods for water ingestion, as described in Appendix A.

Based on the conservative assumption that wine is consumed at the same rate as the average consumption of water (370 L/year or about 1 L/day), the annual dose that corresponds to the highest detected 2000 Livermore Valley tritium value in wine is 15 nSv (1.5 µrem). Assuming a more realistic average wine consumption (52 L/year or 1 L/week of wine) and the mean tritium values from the three sampling areas, the annual doses from Livermore, European, and California wines would be 1.6 nSv (0.16 µrem), 1.0 nSv (0.10 µrem), and 0.40 nSv (0.040 µrem), respectively.

The potential ingestion dose from all foodstuffs grown near the Livermore site for 2000 was realistically below 120 nSv (0.12 µSv or 0.012 mrem). This estimate is as high or higher than dose estimates calculated using other assumptions (see Appendix A). This estimate is a factor of 25,000 lower than an annual background dose (~3000 µSv

or 300 mrem) and a factor of 800 lower than the dose from a typical chest x-ray (100 µSv or 10 mrem) (Shleien and Terpilak 1984). Therefore, although tritium levels are elevated slightly near the Livermore site, doses from tritium ingestion are negligible.

Site 300

In general, LLNL impacts on tritium concentrations in vegetation at Site 300 for 2000 were insignificant. With the exception of vegetation from previously identified sites of contamination, the tritium levels at Site 300 were at or near the limits of detection and comparable to those observed in previous years. The areas where tritium is known to be present in the subsurface soil are well delineated and localized.

The calculated maximum potential annual ingestion dose from vegetation at sampling location DWS, based on the maximum value of 1300 Bq/L (35,000 pCi/L), is 6.2 µSv (0.62 mrem). This dose, based on the conservative modeling assumptions described above, is theoretical—but nevertheless small— because vegetation at Site 300 is not ingested either by people or by livestock.